Shear band formation and mechanical properties of cold-rolled bulk metallic glass and metallic glass matrix composite

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The effects of cold rolling on the mechanical properties of monolithic Vitreloy 1 ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$) BMG and ductile phase reinforced *in-situ* composite ($Zr_{56.3}Ti_{13.8}Cu_{6.9}Ni_{5.6}Nb_{5.0}Be_{12.5}$) have been investigated. The bend strength of the as-cast composite was lower than that of the as-cast monolithic BMG. However, the bend deflection of the as-cast composite (~1.0 mm) was significantly higher than that of the as-cast monolithic BMG. The ductility of the monolithic BMGs is improved by cold rolling. In contrast, the ductility of the metallic glass matrix composite is deteriorated after cold rolling.

1. Introduction

The fabrication of large size bulk metallic glasses (BMGs) has accelerated the development of various BMG systems [1, 2]. However, due to the brittle nature of monolithic BMGs upon applied loading, a widespread application for structural use has been limited [2]. In order to control the mechanical behavior of BMGs, the fabrication of *in-situ* ductile phase reinforced BMG matrix composite system has been attempted in several systems and exhibited a significant enhancement of toughness due to the incorporated secondary phases [3–5]. Among the BMGs and composite systems, the monolithic Vitreloy 1 (Vit. 1, $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$) and ductile β phase reinforced BMG matrix composite ($Zr_{56.3}Ti_{13.8}Cu_{6.9}Ni_{5.6}$-$Nb_{5.0}Be_{12.5}$) are attractive systems due to a slow cooling rate required for large size BMG formation and controllable mechanical properties by incorporated ductile phase [3].

The mechanical behavior of monolithic Vit. 1 and the composite have been studied intensively [3]. It has been reported that the ductile β phase in the composite deflects shear band propagation and promotes the creation of multiple shear band formation, resulting an improvement of ductility, while the monolithic Vit.1 showed a brittle nature upon mechanical tests [3]. In general, it has been understood that formation of a large number of shear bands is crucial in enhancing the ductility of BMG and/or BMG-based composite systems. When BMG-based composite exhibits large elongation, a large number of shear bands usually have been observed at the surface of the specimen after compression tests [6]. At the same time, since the pre-introduced shear band itself can possibly serve as nucleation site for shear band formation, attempts have been made to improve the ductility of BMGs by conventional cold rolling [7].

It has been reported that during cold rolling process of Zr-based monolithic BMG, a large density of pre-introduced multiple shear bands has been generated and thus the ductility is increased due to pre-introduced shear bands, indicating that the increased amount of pre-introduced multiple shear bands provides an opportunity for relatively homogeneous strain relaxation of BMGs [7]. However, a comparative study for BMG and BMG-based composite upon equivalent applied deformation has not been investigated yet, even though a distinct microstructural evolution of BMG and BMG-composite upon cold rolling can provide a useful insight for controlling mechanical properties and for investigating deformation characteristics associated with shear band formation.

In the present study, we selected monolithic Vit. 1 and β phase reinforced BMG matrix composite. Since

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the matrix composition of the composite is comparable to the monolithic Vit. 1, the effect of ductile phase on structural evolution can clearly emerge upon cold rolling process. Upon increase of the thickness reduction ratio (TRR), the total density of shear bands in Vit. 1 gradually increased, while that of shear bands in the composite was explosively increased when the reduction ratio reached 18% of TRR. Following cold rolling of monolithic BMG and the composite with a variation of TRR, microstructural evolutions and mechanical properties have been examined.

2. Experimental

30 g ingots with a nominal composition of monolithic Vit.1 ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (at%)) and *in-situ* composite ($Zr_{56.3}Ti_{13.8}Cu_{6.9}Ni_{5.6}Nb_{5.0}Be_{12.5}$ (at%)) have been made by arc-melting of elemental components and Be-Cu-Ni alloys in a Ti-gettered Ar atmosphere. In order to fabricate a plate-shaped BMG a self-designed plate-shaping equipment was used: after arc-melting the ingot in an Ar atmosphere, the melt is sandwiched immediately between two water-chilled cupper rods. For the preparation of cold rolling specimen, the BMG and composite plates were cut into a rectangular shape with a dimension of about $6 \times 25 \times 4$ mm (width, length and height).

An electric motor-controlled twin roll (20 cm in diameter) with a constant surface speed of about 10 cm/s was used for the cold-rolling process. The distance between rolls was gradually reduced so that a thickness reduction ratio of about 0.5–1% can be reached for each rolling cycle. The equivalent TRR's of 2, 7.5 and 18% were applied for both Vit.1 and composite to compare shear band formation and bending properties. Following cold rolling process, bending tests were carried out by an Instron—type multitester (Hounsfield H25KT) at a quasi-static mode with an initial strain rate of 1×10^{-4} s^{-1}. The specimens for the bending tests were prepared by cutting and polishing the BMG and composite with a dimension of $2 \times 15 \times 1.5$ mm (width, length and height). The microstructures of the cold-rolled BMG were investigated by SEM (Scanning Electron Microscope, Hitachi S-2700). Phase identifications were performed using XRD (X-ray Diffraction, Rigaku CN2301).

3. Results

Fig. 1 shows the results of XRD obtained from as-cast monolithic Vit.1 and BMG-composite. The XRD pattern obtained from Vit.1 showed a typical amorphous halo at around 38° and the XRD pattern obtained from the BMG composite exhibited sharp peaks from *in-situ* formed crystalline β phase superimposed on the amorphous halo peak, corresponding to the previous reports [3]. SEM back scattered electron (BSE) image inserted in Fig. 1 confirmed the presence of dendritic β phase embedded in the amorphous matrix.

The SEM images in Fig. 2a–c show the side views of the cold rolled BMG with TRRs of (a) 2, (b) 7.5 and (c) 18%, respectively. Two distinct types of shear

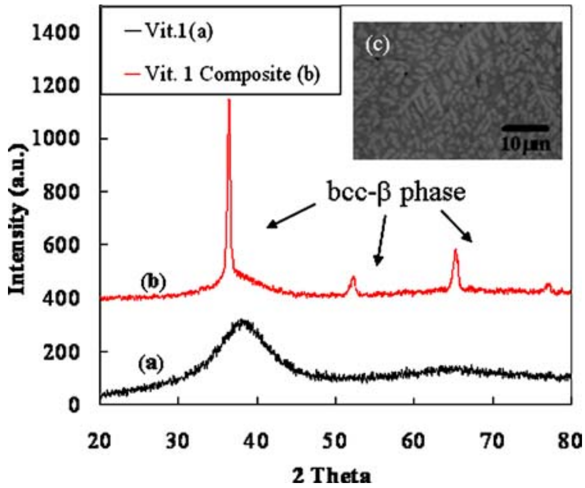

Figure 1 XRD results of (a) vitreloy1, (b) composite and (c) the microstructure of the composite.

bands were developed: primary shear bands inclined about 45° to the rolling direction and secondary shear bands stemmed from the primary shear bands almost in parallel to the rolling direction. The primary shear bands propagated straight passing through whole thickness of the rolled sample. It is specially noted that the density of primary shear bands did not increase significantly with the increase of reduction ratio, while that of secondary shear bands increased with an increase of TRR.

The SEM images in Fig. 3a–c show the side views of the cold rolled composite with TRRs of (a) 2, (b) 7.5 and (c) 18%, respectively. As shown in Fig. 3a, a large number of shear bands started to develop at TRR of 2%. With an increase of TRR, the number of shear bands significantly increased, as shown in the magnified image of Fig. 3d (TRR: 7.5%). For the case of composite, it was difficult to differentiate primary and secondary shear bands. Due to the presence of β phase, the shear bands did not propagate in a straight way, but in an irregular and wavy fashion. The number of shear bands further increased with increasing TRR up to 18%, as shown in the magnified image of Fig. 3e (TRR: 18%). The BSE image in Fig. 3f shows that the shear bands propagate even inside the β phase at 18% TRR.

Fig. 4 shows the bend stress—deflection curve of the as-cast and cold rolled monolithic BMGs. The bending strength was calculated using $\sigma = (3Pl)/(2wt^2)$, where P, load and l, w and t: length, width and thickness of the beam. The as-cast Vit. 1 showed no bend deflection, while the cold rolled Vit. 1 exhibited a deflection up to ~1 mm. However, the deflection did not continuously increase in proportional to the thickness reduction ratio. When the TRR was at between 7.5 and 18%, the bend strength and deflection showed a similar value, indicating that the increased number of shear bands do not dominantly affect the toughness of the specimen at this range of TRR.

Fig. 5 shows the results of bending tests of the as-cast and cold rolled composite samples. Since the ductile β phase is incorporated in the amorphous matrix, the bend strength of the as-cast composite was lower than that

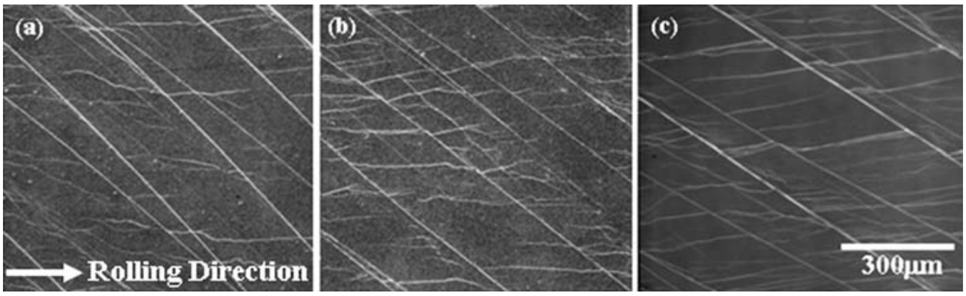

Figure 2 SEM of monolithic vitreloy1 with a thickness reduction ratio of (a) 2%, (b) 7.5% and (c) 18%.

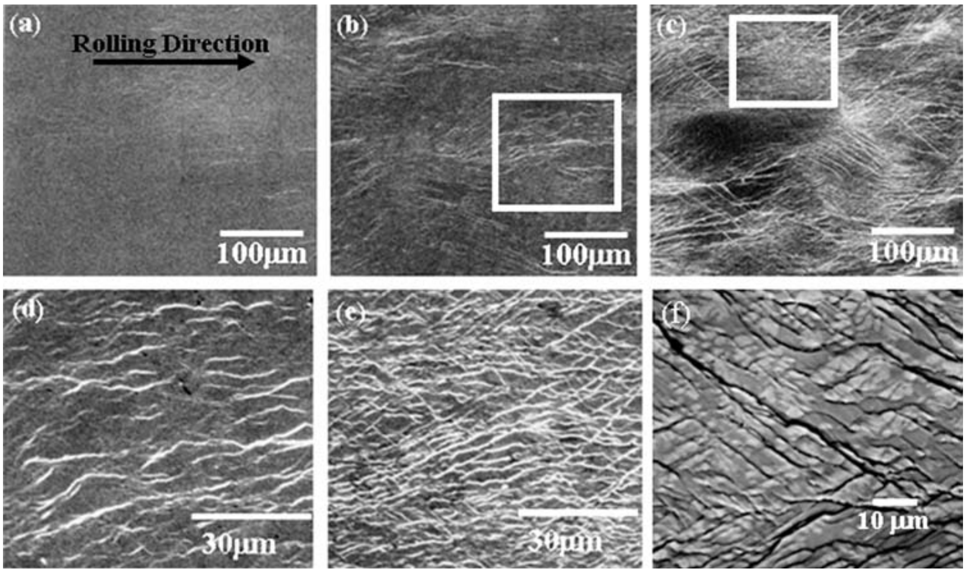

Figure 3 SEM of the composite with a thickness reduction ratio of (a) 2%, (b) 7.5%, (c) 18% and (f) BSE image of the composite with 18% reduction ratio ((d) and (e) is the enlargement of (b) and (c), respectively).

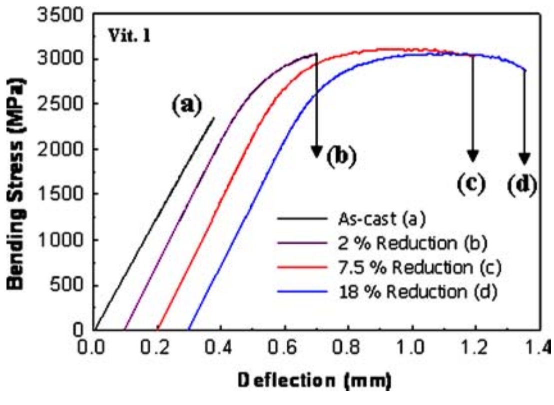

Figure 4 Bend strength—deflection curve of monolithic Vitreloy 1 of (a) as-cast and with a thickness reduction ratios of (b) 2%, (c) 7.5% and (d) 18%.

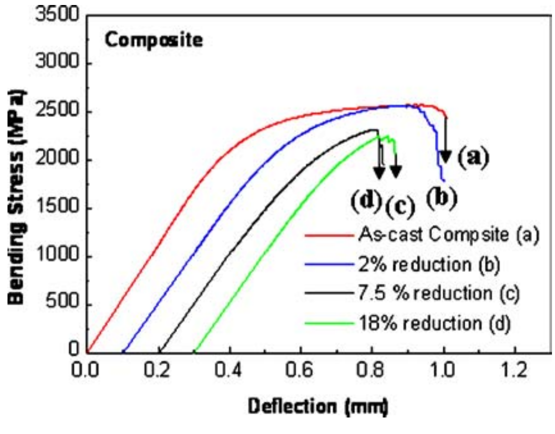

Figure 5 Bend strength—deflection curve of the composite specimens of (a) as-cast and with a thickness reduction ratios of (b) 2%, (c) 7.5% and (d) 18%.

of the as-cast monolithic BMG. However, the bend deflection of the as-cast composite (∼1.0 mm) was significantly higher than that of the as-cast monolithic BMG. The bend strength and deflection of the composite decreased with increasing TRR, i.e. the bend strength of as-cast and 18% cold rolled samples were 2600 and 2300 MPa, respectively, and the bend deflection of the as-cast and 18% cold rolled samples were 1.0 and 0.6 mm, respectively. In contrast to the case of monolithic BMG, the bend deflection of the composite decreased significantly with cold rolling. When the composite was cold-rolled up to 18% of TRR, the total deflection was about lowered by about 40%.

4. Discussion

Present results show that cold rolling is one of the useful processes that can provide shear bands in monolithic BMG and possibly can lead to the enhancement of plasticity, when an appropriate amount of TRR is

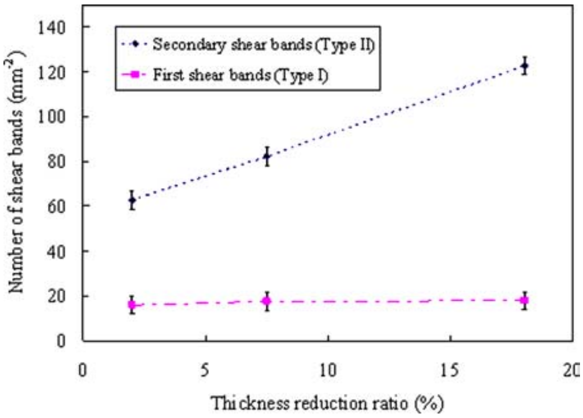

Figure 6 Shear band density of monolithic vitreloy1 with a variation of thickness reduction ratios.

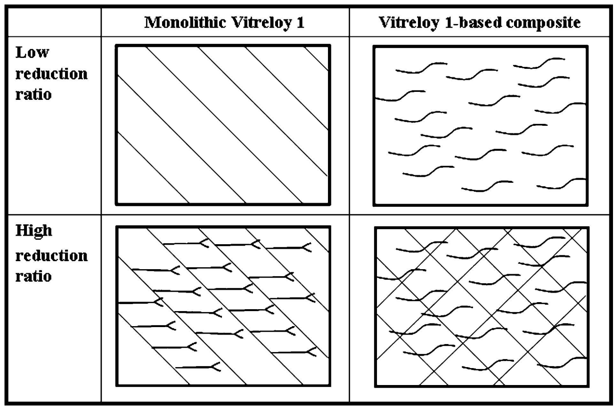

Figure 7 Schematics of shear band development during cold rolling process.

applied. The density of shear bands developed during cold rolling of monolithic Vit. 1 has been estimated at TTRs of 2, 7.5 and 18% (Fig. 6). The number of the primary shear bands is almost constant with the increase of TRR. However, the secondary shear bands density increases significantly with increasing TRR, i.e. about a factor of two times when TRR is increased from 2% to 18%. It appears that the density of the primary shear bands is saturated very fast at a low level of applied deformation. When TRR is increased further, only the density of the secondary shear bands increases, implying that the secondary shear bands provides a main stress relaxation site during deformation. It should be noted that since the ductility of BMG is mainly affected by the shear band formation, the present observations clearly show that the limited ductility of monolithic BMG is due to the limited number of primary shear band formation, which leads to a catastrophic fracture upon applied loading. For the case of cold rolling of Zr-Cu-Al-Ni amorphous alloy, the density of shear bands has been reported to be about 250 per mm^{-2} at about 20% in thickness reduction [7]. However, the present study shows that the density of the shear bands is about 140 per mm^{-2} at 18% in thickness reduction, which is lower than the previously reported value, inferring that the shear band density developed during cold rolling may be dependent on the compositional variation and/or processing parameters such as thickness reduction ratio for each pass and initial thickness of the sample.

On the contrary, for the case of the composites, the primary and secondary shear band can not be differentiated easily, since most of the shear bands initiate from the interface between the β phase and amorphous matrix. When the stress is applied during cold-rolling, the β phase will deform first. But the deformation is strictly confined due to the surrounding amorphous matrix (i.e. at the interface). At the same time, the specimen undergoes a thickness reduction upon cold rolling, implying that the plastic deformation affected the β phase and amorphous matrix (the composite passes over elastic limit). In fact, the β phase and amorphous matrix deform in a different manner. The ductile β phase undergoes relatively homogenous deformation but the amorphous matrix experiences inhomogeneous deformation, which provides shear bands formation in the matrix. Since the propagation of shear bands in composite samples is blocked by the β phase, the shear bands can not propagate in a straight way, but in an irregular manner mostly along the interface. The irregular shape of the shear bands are shown in back scattering image of Fig. 3f. It clearly shows that the shear bands are by-passed and redirected by the incorporated ductile phase, leading that the shear bands are not really a linear shape. The reason for the development of shear bands in a wavy fashion is considered as (a) a large number of the shear band formation at the interface between ductile phase and amorphous matrix and (b) shear bands are by-passed along the ductile phase (c) a multiple rolling with a small reduction ratio can continuously provide nucleation site of the shear bands and the interaction of the shear bands. The estimation of shear band density clearly shows a large increase of shear bands with increasing TRR. The density of the shear bands at 18% of TRR is increased by about two orders when compared with the monolithic BMG at the same TRR.

As previously reported [7], cold rolling of monolithic BMGs leads to the enhancement of ductility. However, the ductility of the metallic glass matrix composite is deteriorated after cold rolling. The bending deflection of the composite decreases significantly, when TRR is over 7.5% (Fig. 5). When the as-cast composite is deformed upon loading the ductility of the ductile beta phase reinforced composite is much larger than monolithic BMG, since the ductile β phase can block and redirect shear band propagation (ductile phase toughening) [3]. However, for the present observations, the cold rolled composite did not show enhanced ductility. In principle, if the ductile β phase undergoes work hardening upon cold rolling, the β phase should act as an effective barrier for blocking shear band propagation. In this regard, it is probable that the ductile phase would undergo work hardening and the deformed ductile phase can not serve any more as an effect barrier for shear band propagation. Further structural identification is underway.

At the same time, the bend strength was also lowered for the cold rolled composite. It has been suggested that the atomic free volume in shear bands is larger than that in matrix of BMG [8, 9]. The BMG containing pre-introduced shear bands is similar to a ductile

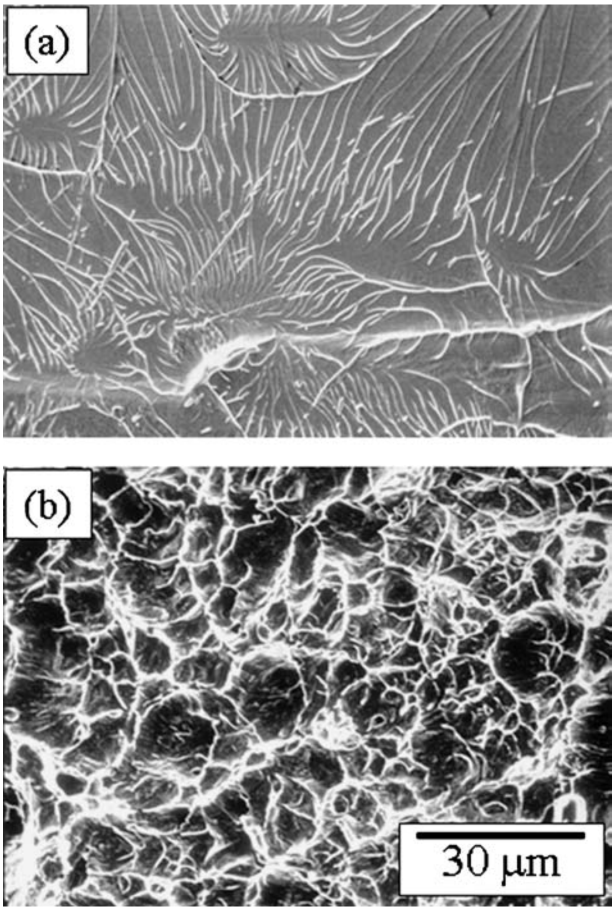

Figure 8 Fractured surface of (a) Vitreloy 1 and (b) composite deformed by cold rolling with a thickness reduction ratio of 7.5%.

phase reinforced composite system, which is composed of hard undeformed regions and soft deformed regions, resulting that the mechanical response would exhibit reduced yield strength but enhanced ductility [7]. Therefore, it seems that the strength drop of the cold rolled composite is due to the enhanced free volume upon deformation [7, 8]. The tensile fractured surface after bending tests (7.5% cold rolled BMG) is shown in Fig. 8a. Typical vein-like patterns that develop in tensile modes were observed. In contrast, the tensile fracture surface for the 7.5% cold rolled composite, showed a ductile dimple fracture, which is a typical indication of the ductile fracture.

5. Summary

The structural evolution of monolithic BMG ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (Vit. 1)) and BMG-based composite ($Zr_{56.3}Ti_{13.8}Cu_{6.9}Ni_{5.6}Nb_{5.0}Be_{12.5}$) has been examined in terms of applied deformation by cold rolling. While the number of the primary shear bands in the monolithic BMG is constant with the increase of TRR, the secondary shear bands density increases significantly with increasing TRR. For the case of the ductile β phase reinforced composites, the shear band density increases dramatically with increasing TRR. The density of the shear bands at 18% of TRR is increased by about two orders when compared with the monolithic BMG at the same TRR. The bend strength of the as-cast composite was lower than that of the as-cast monolithic BMG, while the bend deflection of the as-cast composite ($\sim$1.0 mm) was significantly higher than that of the as-cast monolithic BMG. After cold rolling, the ductility of the monolithic BMGs is improved, while the ductility of the metallic glass matrix composite is deteriorated after cold rolling.

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